The origin of spin current in YIG/nonmagnetic metal multilayers at ferromagnetic resonance*

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Spin pumping in yttrium-iron-garnet (YIG)/nonmagnetic-metal (NM) layer systems under ferromagnetic resonance (FMR) conditions is a popular method of generating spin current in the NM layer. A good understanding of the spin current source is essential in extracting spin Hall angle of the NM and in potential spintronics applications. It is widely believed that spin current is pumped from precessing YIG magnetization into NM layer. Here, by combining microwave absorption and DC-voltage measurements on thin YIG/Pt and YIG/NM_1/NM_2 (NM_1 = Cu or Al, NM_2 = Pt or Ta), we unambiguously showed that spin current in NM, instead of from the precessing YIG magnetization, came from the magnetized NM surface (in contact with thin YIG), either due to the magnetic proximity effect (MPE) or from the inevitable diffused Fe ions from YIG to NM. This conclusion is reached through analyzing the FMR microwave absorption peaks with the DC-voltage peak from the inverse spin Hall effect (ISHE). The voltage signal is attributed to the magnetized NM surface, hardly observed in the conventional FMR experiments, and was greatly amplified when the electrical detection circuit was switched on.

Keywords: spin current, spin pumping, inverse spin Hall effect, magnetic proximity effect, magnetic insulator

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1. Introduction

Spin current generation, detection, and manipulation involve fundamental science as well as the key-technologies[1,2] in spintronics. Spin pumping from precessing magnetization under ferromagnetic resonance (FMR) conditions is an attractive method of generating coherent pure spin current.[3–8] Pure spin current is the key resource in spintronics as well as a base for studying ISHE characterized by the spin Hall angle α_{SH}. Ferromagnetic-insulator (FI)/nonmagnetic-metal (NM) bilayers are believed to be the ideal settings for measuring α_{SH} of the non-magnetic metals. One of the widely studied such systems is yttrium-iron-garnet (YIG)/Pt bilayer[9–16] because YIG [$Y_3Fe_2(FeO_4)_3$] is a well-known insulating magnetic material and Pt is a well-studied heavy metal with a strong spin–orbit interaction. The conventional understanding of the system under FMR conditions is that magnetization of YIG precesses coherently, and the precessing magnetization pumps pure spin current into Pt layer[9–12] across the YIG/Pt interface. The spin current in the Pt layer is then converted into a transverse (normal to spin flow direction and spin polarization) charge current that can be detected as an electrical voltage signal. YIG/Pt bilayer is believed to be a clean system for studying spin pumping and ISHE since YIG cannot conduct the electric current so that whatever the DC-voltage measured in Pt must be from the ISHE. The signature of spin pumping is the broadening of the FMR peak width of YIG as well as a detected DC-voltage in the Pt layer. Controversially, the extracted values of α_{SH} from different groups differ by three orders of magnitude[10,16,17] in this "clean" system. It arises the question of who is responsible for the spin pumping, YIG or other magnetic sources that varies from sample to sample. The answer to the question requires a good understanding of the interfacial phenomena between the YIG and an NM.

In this paper, we use thin YIG(16 nm)/Pt(10 nm) bilayer and

YIG(16 nm)/Cu(5 nm)/Pt(10 nm),

YIG(16 nm)/Cu(5 nm)/Ta(10 nm),

YIG(16 nm)/Al(5 nm)/Pt(10 nm)

trilayer systems in a rectangular cavity to investigate these issues by a combined measurements of microwave absorption

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and DC-voltage. The reason of using such thin YIG films is that only the uniform precessing spin wave can be excited in an in-plane FMR experiment so that one can examine whether the electrical signal is from the spin wave of the YIG. This is in contrast to the thick YIG films,[9] where enormous number of FMR peaks and voltage peaks were excited so that a careful comparison is very difficult, if it is not impossible. Contrary to the popular belief, the spin current in Pt or Ta was not pumped from the precessing YIG magnetization, but from the magnetized NM surface (in contact with thin YIG) originated either from the magnetic proximity effect (MPE)[18–24] or from the inevitable diffused Fe ions from YIG to NM.[25] Interestingly and surprisingly, the FMR signal from the magnetized NM surface was greatly amplified when the electrical measurement circuit was connected (otherwise, the signal could hardly be observed). When the NM is not magnetized, such as in thin YIG(16 nm)/Al(5 nm)/Pt(10 nm) samples, no detectable DC-voltage signal was observed in Pt. Our experiments showed unambiguously that spin pumping from the very thin insulating YIG layer into the metallic Pt or Ta layer was not efficient and effective, in comparison with that from a magnetized metallic surface into Pt or Ta.

2. Our experimental methods

2.1. Sample preparation and experimental procedure

Very thin YIG [$Y_3Fe_2(FeO_4)_3$] films (16 nm) were fabricated on $Gd_3Ga_5O_{12}$ (GGG) wafers by PLD. X-ray diffraction (XRD) and atomic force microscopy (AFM) showed that our YIG are high quality (See Fig. S1). The in-plane FMR spectrum shows only one uniform mode with linewidth about a few Oersteds, and $4\pi M_s$ is about 0.203 T from Kittel formula (See Fig. 1(a) and Fig. S2(a)). These values were very closed to one obtained by Lin *et al.*[26] Py, Pt, Cu, Ta or Al with high purity (4N) was then deposited on YIG by magnetron sputtering to create an NM strip with a mask of 0.2 mm × 2.3 mm. All samples were cut into 1 mm × 3 mm for DC-voltage and microwave absorption measurements in a homemade X-band microwave absorption spectrometer. The microwave power was fixed at 32 mW.

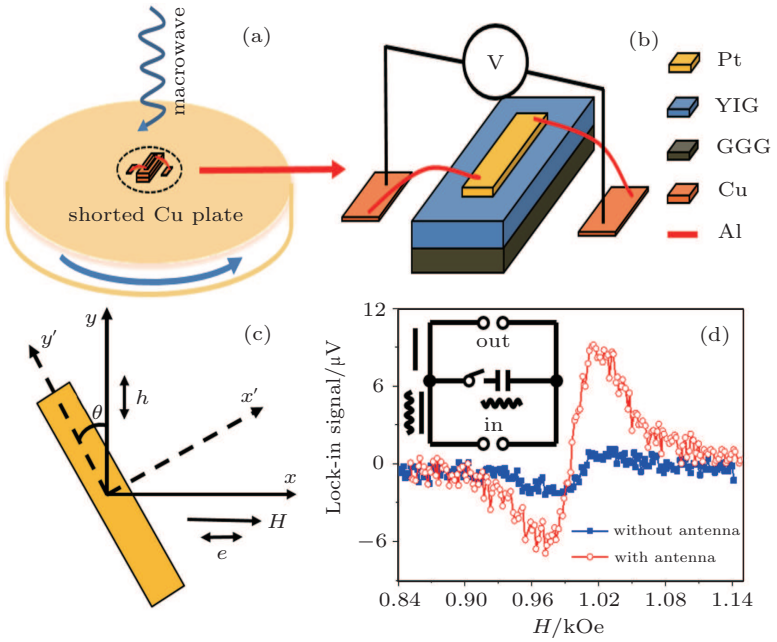


Fig. 1. (color online) (a) The experimental setup for the microwave absorption measurement and electrical detection of FMR in which DC-voltage along the long edge of the sample was measured. (b) The zoom-in of sample and copper-pads. The total thickness of the sample including GGG substrate is about 1 mm. (c) The in-plane rotation geometry of sample. The microwave of frequency $f = 9.7$ GHz propagated along the z axis, and the external field H was along the x axis. e and h are the electric and magnetic components of the microwave, respectively along the x axis and the y axis. The angle between the y axis and the long edge of sample was denoted as θ that varies as the shorted copper plate rotates in the xy-plane. (d) FMR signals of 3-nm Py thin film strip when the Al wires were connected/disconnected to the Cu-pads (with/without antenna effect). The inset is the equivalent circuit with antenna effect.

The experimental setup is shown in Fig. 1. The FMR microwave absorption and the DC-voltage were measured at frequency $f = 9.7$ GHz of TE_{10} mode in the X-band cavity. The sample with size of 1 mm × 3 mm was mounted in the middle of a shorted copper plate at one end of the cavity that can rotate in the xy-plane as illustrated in Fig. 1. The angle between the y axis and the long edge of the sample is denoted as θ. Two thin rectangular copper sheets of 1.5 mm × 8 mm were symmetrically placed on the both sides (1 mm away from the sample) of the sample in the cavity as illustrated in Fig. 1. These two small copper sheets were isolated from the shorted copper plate and acted as electrical connection pads that connected to an SR-530 lock-in amplifier of Stanford Research Systems or Keithley-2182 nanovoltmeter. It should be pointed out that these two thin copper sheets did not affect the X-band microwave distribution from the angular dependence measure-

ments of FMR for Py (see Fig. S3). Two Al wires of diameter about 30 micrometers were attached to two long-edge ends of the sample. As illustrated in Fig. 1, the electrical measurement circuit was switched on when the other ends of Al wires were connected to the two Cu-pads. The in-plane external field and AC microwave field were always orthogonal with each other in order to have a maximal precessing magnetization.

2.2. Angular dependence

The DC-voltage from ISHE, AMR, and/or AHE of a ferromagnetic metal near the FMR has a symmetric Lorentz lineshape and an asymmetric dispersive lineshape[27–29]

$$\begin{aligned} U &= U_{\rm sym}L(H,H_0,\Gamma)+U_{\rm asy}D(H,H_0,\Gamma),\\ L(H,H_0,\Gamma) &= \frac{\Gamma^2}{(H-H_0)^2+\Gamma^2},\\ D(H,H_0,\Gamma) &= \frac{\Gamma(H-H_0)}{(H-H_0)^2+\Gamma^2}, \end{aligned} \tag{1}$$

where, H_0 and Γ are respectively the resonance field and resonant peak width. $U_{\rm sym}$ and $U_{\rm asy}$ are the voltages of the symmetry and asymmetry components of the DC voltage that depend on angle θ as[30]

$$\begin{aligned} U_{\rm sym} &= U^{\rm s}_{\rm SR}\sin(2\theta)\sin(\theta)+U_{\rm SP}\cos(\theta),\\ U_{\rm asy} &= U^{\rm a}_{\rm SR}\sin(2\theta)\sin(\theta), \end{aligned} \tag{2}$$

where $U^{\rm s}_{\rm SR}$ and $U^{\rm a}_{\rm SR}$ are the voltages due to the spin rectification. $U_{\rm sp}$ is the voltage from the ISHE due to spin pumping.

2.3. First-principle calculations

Because our computation resources do not allow us to perform reliable calculations on YIG/Pt and/or YIG/Cu systems due to the huge unit cells, we performed the calculations on Ni (111)/Cu and Ni (111)/Pt to analogize YIG/Cu and YIG/Pt systems using the same method presented in Ref. [19] for calculating MPE of Pt in Ni (111)/Pt systems.

3. Experiment results

3.1. YIG(16 nm)/Pt(10 nm) systems

The black circles in Fig. 2(a) are the derivative microwave absorption spectrum ($\mathrm{d}I/\mathrm{d}H$) of a pure YIG film as a function of external magnetic field H. The lineshape of the FMR derivative absorption spectrum of the pure thin YIG sample follows a standard differential Lorentzian line with FMR peak at $H = 2.497$ kOe (1 Oe = 79.5775 A/m), and peak width of $\Gamma = 10$ Oe which shows high quality of our thin YIG samples.[16] The blue circles in Fig. 2(a) are the FMR derivative absorption spectra of a thin YIG(16 nm)/Pt(10 nm) bilayer strip when the electrical detection circuit is switched off (a conventional FMR configuration used by many groups). As compared with the FMR of the pure YIG film, the FMR derivative microwave absorption spectrum of thin YIG/Pt bilayer sample appears to shift to a lower field with a seemingly broaden peak width that was observed in previous studies[5,16,31] and was used as an essential evidence of spin pumping from YIG. A careful examination showed that the above observed FMR absorption curves of switch-off circuit are better described by two independent FMR signals. One of them with a relative amplitude of $A_1 = 60.5\%$ was from the free YIG because it has the same peak position and peak width of $H_1 = 2.497$ kOe and $\Gamma_1 = 10$ Oe as the free YIG. The other with peak position $H_2 = 2.488$ kOe, peak width $\Gamma_2 = 12$ Oe, and relative amplitude $A_2 = 39.5\%$ was naturally attributed to the YIG covered by Pt. Because Pt modifies magnetic properties of YIG,[31] the peak position and peak width of the second FMR signal differ slightly from those of the free YIG. In fact, our FMR results on pure YIG and YIG(16 nm)/Pt(10 nm) films agree completely with those observed by many other groups.[5,16,31]

The green circles in Fig. 2(a) are the FMR derivative absorption spectra of the thin YIG(16 nm)/Pt(10 nm) bilayer strip when the electrical detection circuit is switched on (see the left inset of Fig. 1(a) and the methods below). Strikingly, as compared with the FMR spectra when the circuit is switched off, the microwave absorption signals were greatly amplified. It is worthy to note that the FMR absorption curves of switch-on circuit were best fitted by three independent FMR signals as shown in Fig. 2(b). Among the three signals in Fig. 2(b), two signals (with $A_1 = 54.5\%$ and $A_2 = 36.4\%$) are exactly those of free YIG and Pt-covered YIG, and the third signal of $H_3 = 2.477$ kOe, $\Gamma_3 = 14$ Oe, and $A_3 = 9.1\%$ came from the amplification of a very weak signal (hidden in the blue circles) originated from the MPE-induced magnetized Pt surface that was in contact with YIG. It agrees with those reports[18–24] that the paramagnetic Pt is magnetized at the interface when it directly contacts with a ferromagnetic material. Furthermore, the corresponding DC-voltage detected in Pt was from the magnetized Pt surface since their peak positions and peak widths match exactly with each other as shown in Fig. 2(b).

To substantiate this interpretation, we fabricated also thin YIG/Pt bilayer samples in which YIG was fully covered by Pt layer. As shown in Fig. 2(c), the blue and green circles are the FMR absorption signals when the electrical measurement circuit was switched on (green) and off (blue). As expected, the signal from the free YIG was absent. In Fig. 2(d), the FMR absorption curves of switch-on circuit now consist of two signals respectively from the Pt-covered YIG and the magnetized Pt surface. Again, the DC-voltage relates to the signal of the magnetized Pt surface since their peak positions and widths match exactly with each other, and cannot be from the spin pumping of thin YIG.

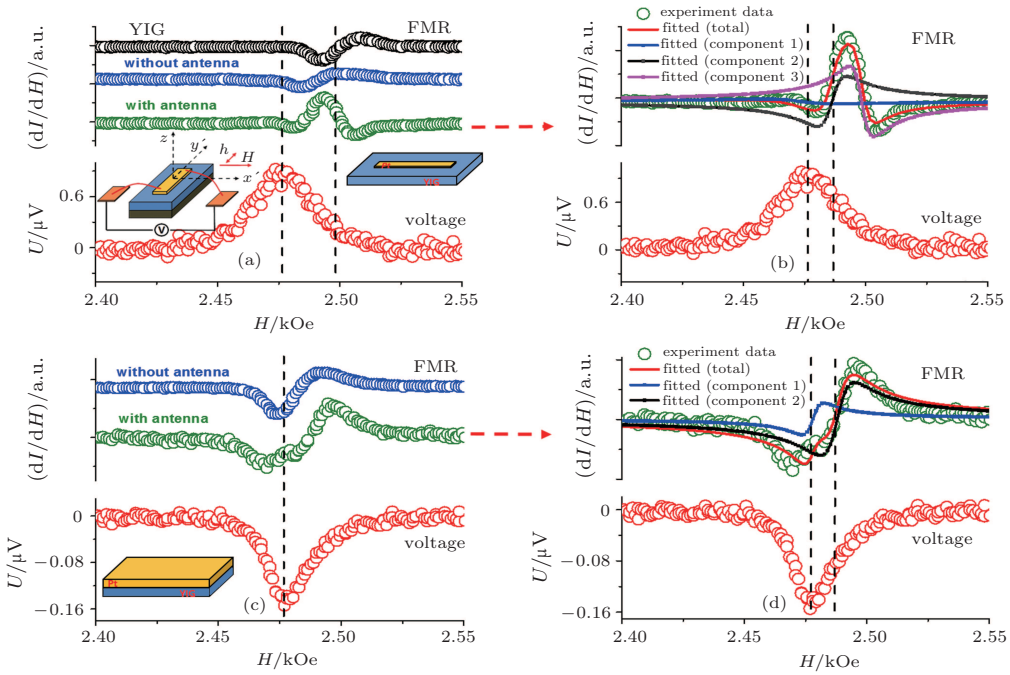


Fig. 2. (color online) (a) The FMR derivative absorption spectra of free YIG and YIG/Pt bilayer strip with $\theta = 0^\circ$ (θ is the angle between microwave field and the sample long edges). The lower panel is the corresponding DC-voltage (bottom) spectra of YIG/Pt bilayer schematically illustrated on the right. The left inset is the schematic diagram of the electrical. The right inset shows the sample structure YIG/Pt stripe. (b) The fit of FMR spectrum of YIG/Pt strip obtained with antenna effect by three FMR signals respectively for free YIG, YIG covered with Pt, and magnetized Pt surface in contact with YIG. The DC signal agrees with the assertion of spin pumping from the magnetized Pt surface. (c) The FMR derivative microwave absorption (upper) and DC-voltage (lower) spectra of fully covered YIG/Pt bilayer (illustrated in the lower left) with $\theta = 170^\circ$ (field was reversed so that the voltage polarity is changed). The inset shows the YIG sample fully covered by Pt. (d) The FMR spectrum of fully covered YIG/Pt bilayer with the antenna effect is best fitted by two FMR signals from YIG covered with Pt and magnetized Pt surface. The corresponding DC-voltage signal was from the magnetized Pt surface. The unit a.u. is short for arbitrary unit.

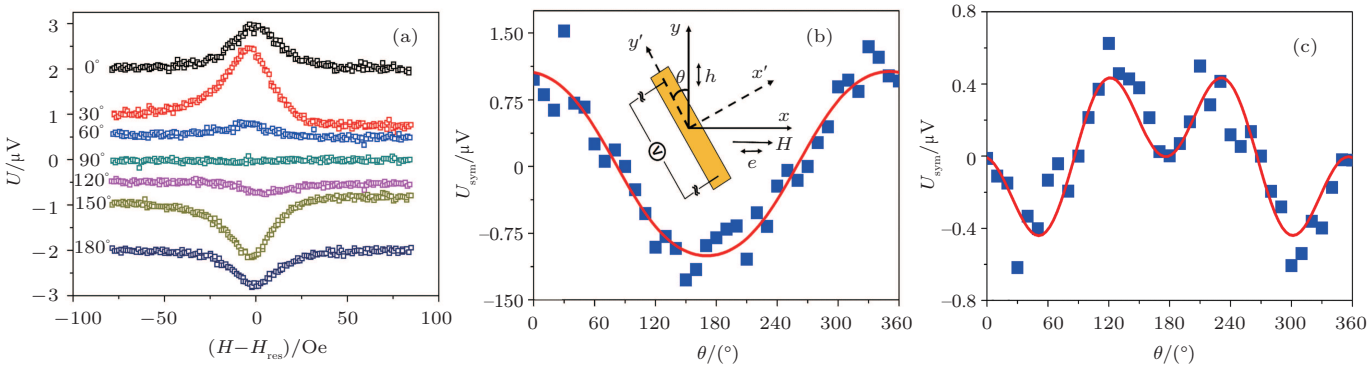


Fig. 3. (color online) (a) The DC-voltage spectra of a YIG(16 nm)/Pt(10 nm) strip at various angle θ. The symmetrical (b) and asymmetrical (c) components of DC-voltages defined in Eq. (1) were extracted. The solid lines are the best fits of Eq. (2) with $U_{\rm SR}^{\rm s} = 0.065$ μV, $U_{\rm SR}^{\rm a} = 0.568$ μV, and $U_{\rm sp} = 1.02$ μV. The inset illustrates experimental setup and angle θ.

The above conclusion could also be reached from the change of the shape of voltage-H curves as angle θ between AC-magnetic field and sample long edge varies. The DC-voltage originated from the spin pumping of YIG should follow a Lorentzian lineshape since the FMR absorption is described by the Lorentzian function.[31] Thus the DC-voltage lineshape would be symmetric about its peak for any angle θ if the spin pumping was from YIG. However, as plotted in Fig. 3(a), it is clear that most spectra consist of a superposition of a Lorentz- and a dispersive-type resonance lineshape.[27,28]

For a given θ, the DC-voltage curve was fitted to Eq. (1) so that both symmetric and asymmetric components of the DC-voltages $U_{\rm sym}$ and $U_{\rm asy}$ were obtained. Their angle-dependences were plotted in Fig.3(b) that fit well with theoretical prediction of Eq. (2) (see the methods below). Voltages $U_{\rm SR}^{\rm s}$ (for symmetric component) and $U_{\rm SR}^{\rm a}$ (for asymmetric component) due to the spin rectification are $U_{\rm SR}^{\rm s} = 0.065$ μV and $U_{\rm SR}^{\rm a} = 0.568$ μV. The voltage $U_{\rm sp}$ from the ISHE due to spin pumping is $U_{\rm sp} = 1.02$ μV. Thus it shows that substantial amount of the DC-voltage came from the AMR and AHE of a ferromagnetic metal that resulted in an asymmetric lineshape, and the natural conclusion is that the Pt surface in contact with YIG was magnetized and generated a spin rectification voltage.[29] The DC-voltage signal from the spin pumping is larger than that from spin rectification, and this magnitude cannot be from spin current pumped from the uniform preces-

sion of the YIG magnetization as shown in a simple estimate in the supplementary materials (see Appendix A). These results further confirmed that the electrical signal is from the magnetized Pt surface in our thin YIG/Pt system.

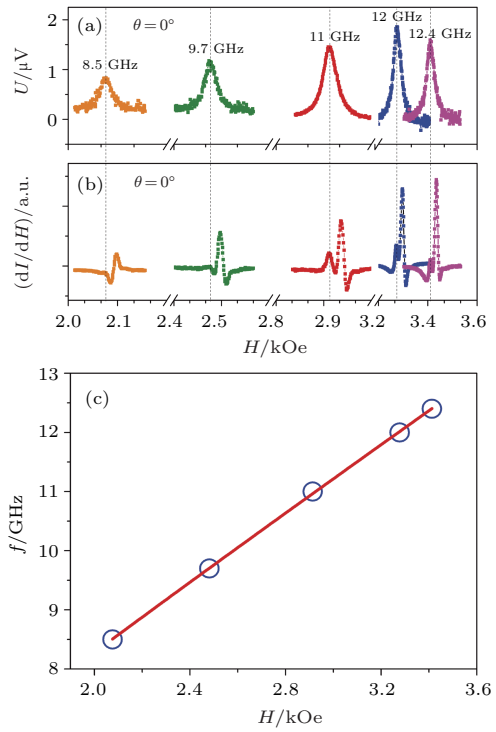


Fig. 4. (color online) The H-dependence of the DC-voltage (a) and FMR spectra (b) of YIG/Pt stripe line at $\theta = 0°$ and for various frequencies with antenna effect. (c) The frequency dependence of peak position of DC-voltages. The solid line is the fit to the Kittel formula.

Figure 4 is the H-dependence of the derivative microwave absorption with the switch-on circuit and the DC voltage of a typical thin YIG/Pt strip sample for various frequencies at $\theta = 0°$. The DC voltages (Fig. 4(a)) have a symmetric Lorentzian shape while the microwave absorption has multi-peaks (Fig. 4(b)) due to different FMR sources. This implies that only one FMR source pumped spin current into Pt and generated the DC-voltage. Clearly, the peak position and width of the DC-voltage match well with those of the tiny FMR signal. In order to show that this peak is not from YIG, the saturation magnetization and the gyromagnetic ratio associated with the observed electrical signal were extracted from our experimental data. Figure 4(c) shows the frequency dependence of the peak field of the DC-voltage that fits well with the Kittel formula,[32,33] $f = (\gamma/2\pi)(H_{\rm res}(H_{\rm res} + 4\pi M_{\rm s}))^{1/2}$, with the gyromagnetic ratio $\gamma = g\mu_{\rm B}/h = 1.738 \times 10^{11}$ rad$\cdot$T$^{-1}\cdot$s^{-1}, slightly away from the known YIG value of 1.785×10^{11} rad$\cdot$T$^{-1}\cdot$s^{-1}, and the saturation magnetization $M_{\rm s} = 0.248$ T. It gave a Lander factor $g = 2.08$ for magnetized Pt surface. These values are different from $M_{\rm s} = 0.200$ T for typical thin YIG grown by pulsed laser deposition (PLD) method due to the deficiency of Fe ions.[41] Our experimental value of $M_{\rm s}$ for MPE of Pt layer in YIG/Pt system, slightly larger probably because the induced moment of Pt in YIG/Pt is slightly higher than that in Ni/Pt since Fe^{3+} in YIG has 5 $\mu_{\rm B}$, is comparable with the reported $M_{\rm s} = 0.224$ T in Ni/Pt by XMCD measurement.[19] Thus, this result further supports our assertion that the precessing magnetization of thin YIG did not pump spin current to Pt layer, contrary to the popular belief.[5,14–16,34]

3.2. YIG(16 nm)/Cu(5 nm)/Pt(10 nm) and YIG(16 nm)/Cu(5 nm)/Ta(10 nm) systems

To further substantiate our claim that the DC-voltage in thin YIG/Pt bilayer is due to the spin pumping from the MPE-induced magnetized Pt layer, a 5-nm thick Cu was inserted between YIG and Pt (Ta) so that Pt (Ta) surfaces were not in contact with YIG and no MPE is possible for Pt (Ta). The upper panel of Fig. 5 is the typical FMR derivative microwave absorption spectrum of one of our thin YIG/Cu/Pt samples. The blue circles are the results when the electrical detection circuit was switched off while the green circles are those when the circuit was switched on. The signal from the magnetized Pt surface was obviously absent, and was replaced by a new FMR signal at an even lower field of $H = 2.46$ kOe, far below YIG resonance peak, and with a peak width of $\Gamma = 12$ Oe. Although this new signal was seen in a 50-times enlarged figure as shown by the black circles in the top panel, it was extremely weak when the electrical detection circuit was switched off. Similar to thin YIG/Pt bilayer samples, an extra signal can be clearly observed when the electrical detection circuit was switched on. As expected this time, the DC voltage of thin YIG/Cu/Pt and/or YIG/Cu/Ta (middle and lower panels of Fig. 5) was observed at $H = 2.46$ kOe, exactly corresponding to the new FMR signal. The peak widths of the FMR and DC-voltage signals matched again with each other as shown in the middle (for YIG/Cu/Pt) and lower (for YIG/Cu/Ta) panels. In contrast, there were no DC-voltage signals at the YIG resonant fields, confirming that the DC-voltage was not due to the spin pumping of YIG, but due to that of the magnetized Cu surface (in contact with YIG).[35–38] Moreover, the signs of the DC-voltages of thin YIG/Cu/Pt and YIG/Cu/Ta samples are opposite due to the opposite sign of spin Hall angle for Pt (positive) and Ta (negative).[16]

Again, above experiments do not support the general belief that precessing YIG magnetization at FMR pumps spin current into Pt or Ta in thin YIG/Pt, YIG/Cu/Pt, and YIG/Cu/Ta systems. Our results are consistent with the assertion that it was the magnetized NM surface pumping spin current into the Pt or Ta layer. The clear evidences include 1) DC-voltage peaks were far from the FMR peaks of the YIG, but were exactly overlapped with the FMR peak of MPE-induced magnetized NM surface; 2) the angle-dependence of the DC-voltage lineshape that shows big contribution from AMR and AHE of a magnetized metal. Furthermore, the MPE of both Pt and Cu were confirmed by our first-principle calculations

(see the Methods). It was found that a few Cu atomic layers adjacent to Ni was magnetized with average moment about $-0.02\ \mu_B$/atom, which was only about 1/5 of the average moment of Pt ($0.11\ \mu_B$/atom) in Ni (111)/Pt system.[19] If one assumes that Cu/Ni and Cu/YIG (Pt/Ni and Pt/YIG) have the similar MPE, then the precession of this small moment can pump spin current into Pt or Ta layer. We can naturally interpret our observed DC-voltage as the ISHE. This smaller magnetized Cu moment explains the much smaller voltage than that in thin YIG/Pt system as shown in Fig. 2(a). The small negative Cu magnetic moment is also consistent with lower resonance field for the magnetized Cu due to the negative exchange field at FMR.[32]

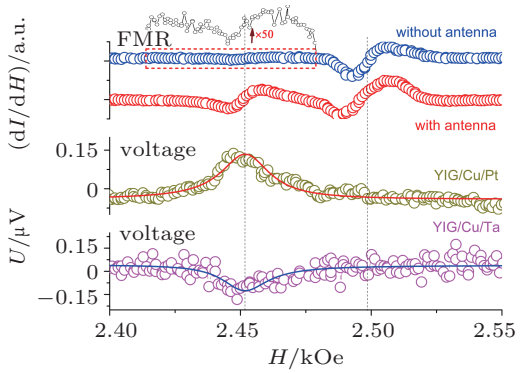


Fig. 5. (color online) The FMR spectra of a YIG/Cu bilayer sample (upper panel) and DC-voltage spectra of YIG/Cu/Pt (middle panel) and YIG/Cu/Ta (lower panel) with $\theta = 0°$. In the upper panel, the green (blue) circles are the FMR derivative microwave absorption spectra when the electrical detection circuit is switched on (off). A weak signal, as shown by the black circles that is the zoom-in (50 times enlarged) of the blue circles inside the red rectangle, was amplified when the electrical circuit was switched on. The DC-voltage in Pt (middle panel) and Ta (lower panel) can be fitted well by the Lorentzian function (solid lines).

3.3. YIG(16 nm)/Al(5 nm)/Pt(10 nm) systems

To further verify the above assertion, we did a controlled experiment with thin YIG(16 nm)/Al(5 nm)/Pt(10 nm) samples. It is clear that the thin YIG(16 nm)/Al(5 nm)/Pt(10 nm) samples only show one FMR peak as shown in Fig. 6, which is almost the same as that of pure YIG in Fig. 2(a). Moreover, the derivative microwave absorption spectrum of a typical thin YIG/Al/Pt samples in Fig. 6 does not change whether the electrical detection circuit was switched on or off, in contrast to that for YIG/Pt or YIG/Cu/Pt(Ta) systems. Especially, no detectable DC-voltage was observed as shown in the bottom panel of Fig. 6.

According to our first-principle calculations on Ni/Al system, there is no significant spin polarization in Al in YIG/Al/Pt system, and correspondingly there is no additional FMR peak from the magnetized Al layer. No observable DC-voltage in Fig. 6 indicates that the FMR of the YIG layer could be hard to pump a detectable spin current into Al/Pt layers in YIG/Al/Pt system efficiently. Therefore, the controlled experiments with YIG(16 nm)/Al(5 nm)/Pt(10 nm) further reveal that spin current in NM layer(s) was not from YIG in thin YIG/NM_1 bilayer or YIG/NM_1/NM_2 multilayer samples, but from magnetized NM1 surface (in contact with YIG).

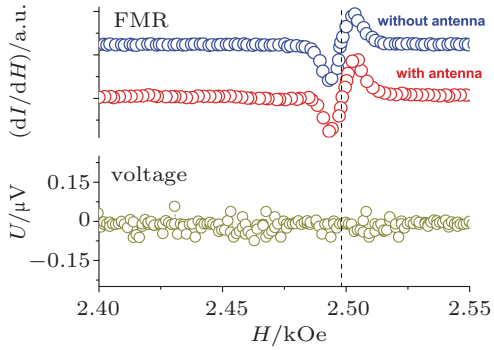


Fig. 6. (color online) Top: The FMR derivative microwave absorption spectra of a YIG/Al(5 nm)/Pt(10 nm) sample when the electrical detection circuit was switched on (red) and off (blue). No MPE-induced magnetized Al was observed. Bottom: No DC-voltage signal in Pt was observed.

4. Discussion

One can make several remarks about spin pumping based on our experiments on very thin YIG films. (i) The electrical signals contain both the contributions from the anisotropic magnetoresistance (AMR) and anomalous Hall effect (AHE). They should be absent if there exists no a magnetized Pt layer. The spin pumping from YIG should not generate such an AMR and AHE signals. The angular dependence of electrical signals agrees neither with spin Hall magnetoresistance. One may argue that these signals are due to the YIG-generated magnetic field effect on non-magnetized Pt. But, there is no any reports of this type of effects so far although such systems have been extensively studied for a long time. (ii) If the unknown signal is due to another spin wave mode of YIG, there is no reason to believe that only the unknown FMR signal can be amplified by antenna effect, but the known uniform spin wave mode of the YIG cannot. (iii) If the unknown FMR peak (as well as the electrical signal) is due to the spin pumping of YIG, then one should expect the same spin wave mode can pump spin current into Pt in YIG/Cu/Pt and YIG/Al/Pt systems with the same resonance field and the same peak width. However, we saw that the peak properties are totally different for YIG/Cu/Pt systems, and no signal for YIG/Al/Pt systems. Moreover, the shift between the resonant field of YIG cover with Pt and dc signal is the same for both partially and fully covered samples (Figs. 2(b) and 2(d). In summary, there would be more problems to associate what were observed in our experiments to spin pumping from YIG than that from a magnetized Pt layer.

We would also like to discuss several possible issues in our experiments. The first issue is the amplification of FMR signal by the electrical-signal detecting circuit. A ferromagnetic metallic film at its FMR can generate not only a DC

signal but also a radio-frequency AC field[29,39] by the AHE and the AMR. As illustrated in Fig. 1 (see the methods) when the sample is connected to Cu connection-pads through two Al wires, the whole structure becomes a patch antenna and a high frequency pass filter. According to the patch antenna theory,[26,40] the AC signals from the AMR and AHE of the ferromagnetic metallic layer and from ISHE of nonmagnetic metallic layer can be radiated through fringing fields at the radiating edges at the same FMR frequency. This will result in the amplification of the FMR signal of the metallic film. Here we termed this amplitude amplification as "antenna effect" when the Al wires were connected to Cu-Pads (see experimental setup, switch-on circuit). Figure 1(d) shows clearly this antenna effect since the microwave absorption of a 3-nm-thick Permalloy (Py ferromagnetic metal) film was greatly enhanced when Al wires were connected to Cu-Pads. Moreover, the antenna amplification effect on ferromagnetic metallic layers does not change the ferromagnetic resonance field and its peak width, as shown in Fig.1(d).

The second issue is about possible multiple FMR peaks due to inhomogeneity in magnetization and surface anisotropy of YIG. It is clear that our pure YIG and YIG/Al/Pt films is homogeneous since only one narrow FMR peak, corresponding to the uniform magnetization precession (spin wave with zero wavenumber), was observed, as shown in Fig. 2(a) and Fig. 6. Moreover, as mentioned before, our FMR results of pure thin YIG and YIG/Pt films agree completely with results from many other groups[5,16,31,41] who did similar measurement on the YIG films of similar thickness, including single FMR absorption peak for YIG. Therefore, the observable inhomogeneity in magnetization and surface anisotropy of YIG can be excluded in our samples with very thin YIG.

The third issue is the possible contribution from spin wave with none-zero wavenumber.[42] For our very thin YIG/NMs samples, it is unlikely to excite the spin wave when the external magnetic field is applied in the film plane.[43,44] Even if spin wave modes of non-zero wavenumbers is excited in the YIG, these spin wave modes and the uniform FMR mode (zero wavenumber) should be amplified together by the antenna effect. However, no amplification was observed on those FMR peaks that did not contribute to the electrical signals. Therefore, one can exclude the electrical signal is from the spin wave modes of non-zero wavenumbers in YIG.

Finally, we discuss why the precessing YIG could not inject a detectable spin current into Pt layer in very thin YIG/Pt sample. One reason may be zero group velocity of the zero-wavenumber spin wave (uniformly precessing YIG magnetization), resulting in a negligible spin current inside YIG. If one assumes that the spin current should be continuous at the YIG/NM interface, the possible pumped spin current (in NM) from YIG should be even smaller. A substantially overestimated upper limit of possible electrical signal due to spin pumping by uniform spin wave of YIG is given in the supplementary materials. It is clear that our observed DC-voltage is at least three orders of magnitude larger than this limit. Another possible reason may be the mismatch between insulating YIG and metallic Pt layers, resulting in an inefficient angular momentum transfer. The FMR linewidth broadening and additional damping mechanism observed previously may be due to the overlap of resonant peaks of both YIG and magnetized Pt.[12,14] Thus one should extract the spin Hall angle α_{SH} by taking into the account of the new findings reported here. We should also emphasis that the present results are based on very thin YIG films, where only one uniform mode of FMR could be found with applied field in the film plane. However, for very thick YIG samples, there might be some spin wave modes that can directly pump spin current into NM layers.[41,45–47]

5. Conclusion

In conclusion, our experiments on very thin YIG/Pt bilayer and YIG/Cu/Pt (YIG/Cu/Ta) trilayer samples showed that the FMR microwave absorption was mainly from three sources: free YIG, YIG covered by an NM, and the magnetized NM surface arising from the MPE. Interestingly, the FMR microwave absorption signal from the magnetized NM layer was pronounced only when the electrical detection circuit was switched on. The electrical detection circuit acted as an antenna for the FMR signal of the magnetized NM surface. Surprisingly, the DC-voltages were from the spin rectification effects and spin pumping of the magnetized NM layers, instead of spin pumping of YIG alone. Thus, contrary to the popular belief, our studies suggest that precessing magnetization of YIG does not pump detectable spin current into the NM layer in very thin YIG/NMs samples. Our findings are very important for properly extracting the spin Hall angle and for a better understanding of the concept of interface mixing conductance.[9–11,16]

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Appendix A: Supplementary information

A1. Characterization of YIG film

The structural quality of our YIG films deposited on (111)-oriented $Gd_3Ga_5O_{12}$ (GGG) substrates by PLD was

characterized by x-ray diffraction (XRD). A representative θ–2θ scan of a 16-nm YIG film shown in Fig. S(1a) exhibits the epitaxial structure with (111) orientation corresponding to GGG substrate. The atomic force microscopy (AFM) image in Fig. S(1b) reveals a smooth surface with an average roughness of 0.17 nm, indicating the high quality of YIG films.

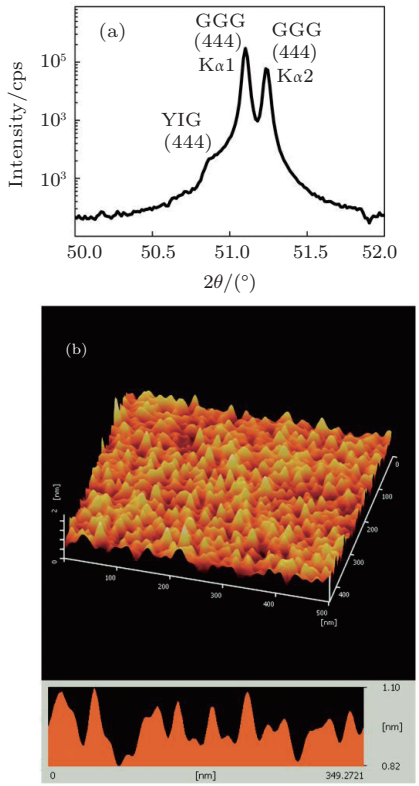


Fig. S1. (color online) (a) The XRD θ–2θ scan and (b) the AFM image of a 16-nm YIG film.

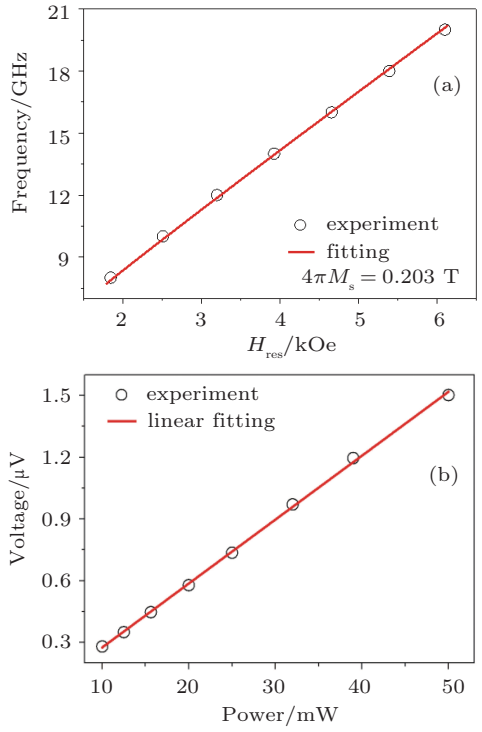


Fig. S2. (color online) (a) Frequency versus resonant field of YIG, the solid line is theoretic fitting based on Kittel formula. (b) The DC voltage as a function of microwave power for YIG/Pt.

The linewidth of FMR is about 10 Oe and $4\pi M_s$ is about 0.203 T from Kittel formula (see Fig. S(2a)). This value is within the range of reported value in literature for thin YIG samples grown by pulsed laser deposition (PLD) method due to the deficiency of Fe ions. Figure S(2b) shows the DC voltage as a function of microwave power for YIG/Pt. The linear behavior indicates that the asymmetric signal observed in Fig. 2 is not due to the nonlinear dynamics of the magnetization of YIG.

A2. Antenna effect

To confirm that the copper sheets (antenna) in the microwave cavity did not affect the microwave transmission, we performed the angular dependence of FMR for Py thin film with the copper sheets that were disconnected from Al wires. FMR spectra are almost identical to each other with or without the Cu sheets (Fig. S(3a)).

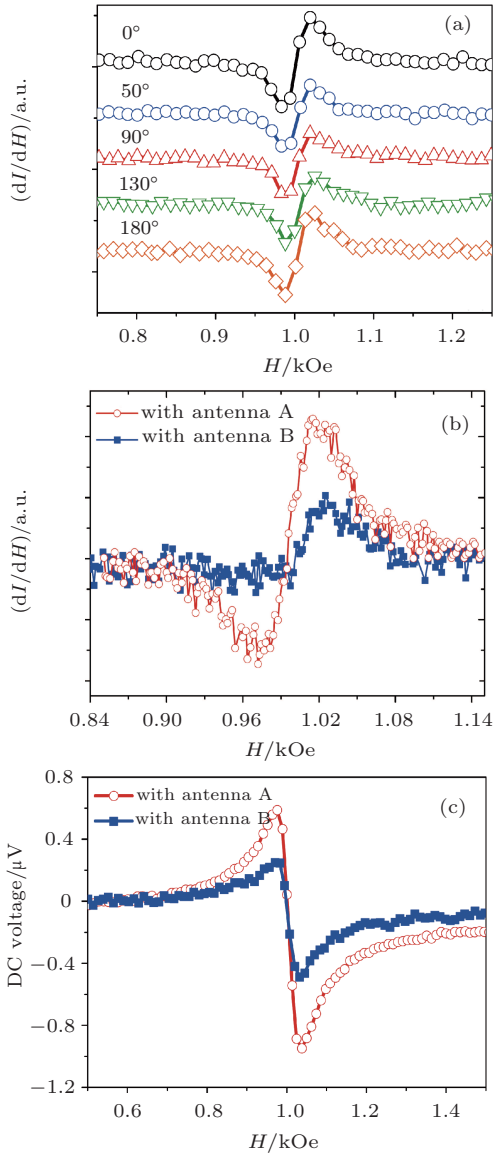


Fig. S3. (color online) (a) Angular dependence of FMR for 10-nm Py thin film for $\theta = 0°$, $\theta = 50°$, $\theta = 90°$, $\theta = 130°$, and $\theta = 180°$. The FMR spectra (b) and the DC-voltage (c) of 3-nm Py strip with resonant antenna A and off-resonant antenna B.

To further confirm the antenna, another antenna of Cu-sheet size of 4 mm × 1.5 mm, termed as off-resonant antenna and denoted as B, is designed. The results with antenna B and with the antenna used in the manuscript (denoted as A whose size is two times bigger than B and termed as resonant antenna) were plotted together in Figs. S(3b) and S(3c). It is clear that the FMR signal (Fig. S(3b)) is more pronounced with resonant antenna A and the DC-voltage (Fig. S(3c)) depends on the antenna effect.

A3. Upper-limit of DC-voltage on Pt by spin pumping from uniformly precessing YIG magnetization

We estimate first the upper-limit of spin current in YIG generated by the uniformly precessing magnetization. The precession angle θ (between the precessing magnetization and spin direction of zero-field) is $\theta = (1/2\alpha)\cdot(b_{\max}/\mu H)$, where α, $b_{\max}$, μ, and H are Gilbert damping coefficient, the maximal RF magnetic field, the magnetic permeability, and the external static FMR resonance field, respectively. The microwave power used in our experiment is $P = 32$ mW and the cross section area of the microwave cavity is $A = 2.2$ cm^2. It gives

$$b_{\max} = \frac{1}{2}\sqrt{\frac{P}{A}\frac{2\mu}{c}} \approx 5.5\times 10^{-7}\ \text{T}.$$

Thus $\theta \approx 10^{-4}$ for $\alpha = 0.0035$[1] and $H = 2.5$ kOe in our experiment. The angle will be even smaller if one takes into account possible magnetic anisotropies of YIG. This means that the number of magnons of zero wave number $q = 0$ cm^{-1} is $n = M(1-\cos\theta)/\mu_{\text{B}} = 1.8\times 10^{20}$ m^{-3} for YIG $M = 1.94\times 10^5$ A/m,[31] where μ_{B} is the Bohr magneton. Thus, the spin current in YIG should be $j_{\text{s}} = (1/2)n\hbar v$, where $1/2$ comes from half of magnons moving toward the interface. v is the group velocity of the magnon that should be zero for $q = 0$ cm^{-1}. However, the linewidth of the FMR is $\Gamma = 12$ Oe at resonance field $H = 2.5$ kOe for resonance frequency $\omega \pm \Delta\omega$. Thus the frequencies of magnons are in the range of $\omega \pm \Delta\omega$, with $\Delta\omega \approx (\Gamma/H)\omega = 0.05$ GHz. As a result, some of the magnons can have non-zero wavenumber whose maximal value is $\hbar\Delta\omega \approx Jq^2$ since the spin wave spectrum is quadratic near $q = 0$ cm^{-1}. For YIG, $J = 1.9\times 10^{-40}$ J$\cdot$m^2. This gives $q = 5.\times 10^4$ cm^{-1} and the group velocity at this wave vector is about $(2J/\hbar)q = 18$ m/s. If one assume that all magnons travel at half of this velocity, Then $j_{\text{s}} \approx 8\times 10^{20}\hbar$ m$^{-2}\cdot$s^{-1}. Of course, this substantially overestimates the spin current because most of magnons do not move at $q = 0$ cm^{-1}. If one assumes that spin current is continuum at the YIG/NM interface and all magnons covert their spins into electron spins in NM, then the DC-voltage at surface of Pt stripe is

$$V = \frac{e}{\hbar/2} j_{\text{s}}\theta_{\text{SH}}\frac{L_{\text{Pt}}}{\sigma_{\text{Pt}}}\frac{l_{\text{sd}}}{d_{\text{Pt}}} \approx 1\ \text{nV},$$

where $\sigma_{\text{Pt}} = 9.7\times 10^6/(\text{m}\cdot\Omega)$, $l_{\text{sd}} = 1.5$ nm,[10,16,17] $L_{\text{Pt}} = 2.3$ mm, and $d_{\text{Pt}} = 10$ nm are the electrical conductivity, spin diffusion length of Pt, the length, and the thickness of Pt strip respectively. In the estimate, we use a large spin Hall angle of $\theta_{\text{SH}} = 0.1$.[10,34]

Even with this substantially overestimated upper-limit of the DC-voltage, our experimentally observed value is still three orders of magnitude larger than this limit. Thus, it is highly unlikely that the uniformly precessing YIG magnetization could pump a large enough spin current into Pt to generated the observed DC-voltage through inverse spin Hall effect.

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